Evolution of biomass burning aerosol properties from an agricultural fire in southern Africa

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[1] Measurements on the UK Met Office C-130 within a distinct biomass burning plume during the Southern AFricAn Regional science Initiative (SAFARI 2000) show an increase in the single scattering albedo as the aerosol ages, from 0.84 at source to 0.90 in the aged regional haze in 5 hours. Condensation of scattering material from the gas phase appears to be the dominant mechanism; the change in black carbon morphology, from a chain to clump like structure, does not significantly affect the bulk aerosol single scattering albedo. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 3359 Meteorology and Atmospheric Dynamics: Radiative processes; 9305 Information Related to Geographic Region: Africa. **Citation:** Abel, S. J., J. M. Haywood, E. J. Highwood, J. Li, and P. R. Buseck, Evolution of biomass burning aerosol properties from an agricultural fire in southern Africa, *Geophys. Res. Lett.*, *30*(15), 1783, doi:10.1029/2003GL017342, 2003.

1. Introduction

[2] Biomass burning aerosol has a large, but poorly quantified radiative effect on climate. The uncertainty in this effect is the result of temporal and spatial variability of the smoke, along with uncertainties in the optical properties of the aerosol itself [*IPCC*, 2001]. *Reid et al.* [1998] found that the aerosol from biomass burning emitted at source had a significantly lower single scattering albedo than the aerosol within the aged regional haze. Quantifying this change in aerosol properties with age is vital if we are to model the climatic effect accurately. This is the first study to analyze measurements of the temporal evolution of biomass burning aerosol single scattering albedo as the smoke is advected downwind from the source.

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2. Instrumentation and Flight Pattern

[3] This study focuses on measurements made by the U.K. Met Office C-130 aircraft during the Southern AFricAn Regional science Initiative (SAFARI 2000) campaign on 13 September 2000 at Otavi in Namibia (flight a790). Sampling was performed over and progressively downwind of a large anthropogenic biomass burning event on agricultural land. The burning had also spread to the surrounding hillsides. The fire consisted of both agricultural and savanna/scrubland fuel sources, with flaming and smoldering combustion occurring.

[4] The C-130 was fitted with the instrumentation detailed in *Haywood et al.* [2003a]. In brief, a Passive Cavity Aerosol Spectrometer Probe (PCASP) was used to measure the aerosol particle distribution from 0.05–1.5 μm radius. A TSI 3563 nephelometer was used to measure the aerosol scattering at 0.45, 0.55, and 0.70 μm. A Radiance Research Particle Soot Absorption Photometer (PSAP) was used to measure the aerosol absorption at 0.567 μm.

[5] The flight consisted of a series of straight and level runs over the source at altitudes ranging from 210–1,233 m above ground level (AGL). This series was followed by a raster pattern of cross plume transits at 1,233 m (standard deviation = 17 m) AGL to 73 km downwind as shown in Figure 1, as is the aerosol number concentration measured with the PCASP. Concentrations up to 100,000 cm^{-3} were observed over the source region, marked with a cross. The two peaks in aerosol number concentration on each side of the cross indicate the areas of most intense burning. The number concentration is shown to spread out and decrease in magnitude as the plume becomes more dilute downwind.

3. Results

3.1. PSAP and Nephelometer Measurements

[6] The single scattering albedo at a wavelength of 0.55 μm, ($\omega_{0\lambda=0.55\ \mu m}$), was calculated along each cross plume transit where the nephelometer scattering coefficient,

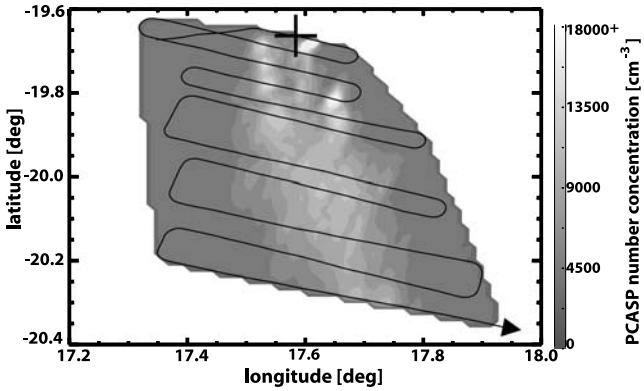

Figure 1. Flight track over and downwind of the fire at Otavi. Measured PCASP number concentration (cm^{-3}) is shown. Source region is marked with a cross.

$\sigma_{s\lambda=0.55\ \mu m}$, exceeded 1×10^{-4} m^{-1}. This limit was the typical background value outside of the influence of the plume, and sharp peaks above this were evident when the aircraft entered the plume itself. The mean atmospheric relative humidity (RH) within the plume where $\omega_{0\lambda=0.55\ \mu m}$ is calculated was 22.1% (standard deviation = 0.7%). The effect of an increase in particle scattering from the nephelometer measurements due to water uptake on the aerosol is negligible as at this low RH [*Magi and Hobbs*, 2003]. The $\omega_{0\lambda=0.55\ \mu m}$ calculation at the source combines measurements from four straight and level runs at various altitudes over the fire (765, 914, 1,057, 1,233 m AGL). Figure 2 shows the evolution of aerosol co-albedo, $(1-\omega_{0\lambda=0.55\ \mu m})$, as the plume moves downwind. The mean and the standard deviation for each cross plume transit are shown. The time subsequent to emission is determined from the measured wind velocity and the aircraft Global Positioning System (GPS). The co-albedo decreases from 0.16 at source to 0.12 two and a half hours downwind, indicating that the aerosol is becoming less absorbing/more scattering as it ages. The standard deviation about the mean is greatest at the source due to the large variability in the smoke plume directly above the fire. *Haywood et al.* [2003a] show that the mean co-albedo of the aged regional haze measured throughout the campaign is 0.10, which suggests that the aerosol measured downwind has not yet fully evolved. An exponential fit suggests it would take approximately 5 hours from emission for the aerosol co-albedo to decrease to 0.10, which is typical of aged regional haze.

3.2. Model Simulations

[7] The observed change in aerosol optical properties subsequent to emission may be the result of changes in the aerosol size distribution, black carbon morphology, chemical composition and the mixing state of the aerosol. These mechanisms are investigated further in 3.2.1 and 3.2.2.

3.2.1. Changes in the Aerosol Size Distribution

[8] $\omega_{0\lambda=0.55\ \mu m}$ was also calculated using the mean PCASP size distributions along each cross plume transit combined with Mie scattering calculations. A refractive index of 1.54−0.018 *i* is used. This is the campaign mean refractive index for the aged regional haze [*Haywood et al.*, 2003a] and is in excellent agreement with AERONET retrieved values on the same day at Etosha Pan [*Haywood et al.*, 2003b]. Etosha Pan is approximately 220 km West from the Otavi fire. The PCASP derived values of co-albedo in Figure 2 are lower than those derived using the PSAP and nephelometer measurements, especially at the source of the aerosol. There is little variation in the modeled co-albedo as the aerosol ages, with a mean value of 0.12 and standard deviation of 0.004. This constancy implies that the aged aerosol effective refractive index is not representative of the refractive index within the plume. Increasing the imaginary part of the refractive index to 0.025 *i* increases the co-albedo at source to 0.16, which is consistent with the independent PSAP and nephelometer result. This result suggests that in this case, it is more important to constrain changes in the effective refractive index than the variation in the size distribution when determining $\omega_{0\lambda=0.55\ \mu m}$ within the plume. However, the Mie scattering calculations assume that the aerosol particles are spherical. *Martins et al.* [1998b] suggest that only smoke particles residing in the atmosphere for more than 1 hour have collapsed sufficiently to be considered spherical. Further investigation into the effect of the variation in particle morphology with age on the aerosol optical properties is made in 3.2.2.

3.2.2. Changes in the Mixing State of the Aerosol and Black Carbon Morphology

[9] Fresh biomass burning aerosol exists as an external mixture in the plume, consisting of quasi-spherical scattering components (organic and inorganic matter) and chain aggregates of black carbon (BC) [*Pósfai et al.*, 2003; *Li et al.*, 2003]. Post combustion BC exists in open chain-like structures of individual spherules. Figure 3 shows a transmission electron microscopy (TEM) image of BC collected in a biomass burning plume by the University of Washington's (UW) CV-580 aircraft in the Timbavati Game Reserve during SAFARI 2000 at a distance between 8−37 km downwind. The exact age of the soot chains are

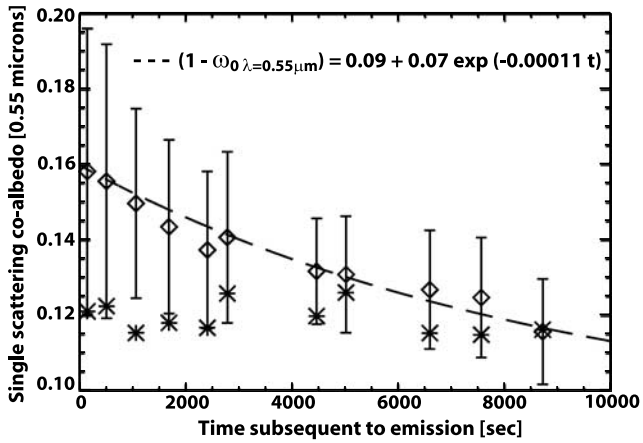

Figure 2. Evolution of the aerosol co-albedo downwind of the fire. Diamonds represent the mean value for each cross plume transit measured with the PSAP and nephelometer. Error bars are the standard deviation from this mean. Values using the PCASP size distribution for each transit and Mie calculations (n = 1.54 − 0.018i) are shown with an asterisk. The exponential fit suggests that it will take 5 hours for the co-albedo to reach 0.10, which is representative of the aged regional haze.

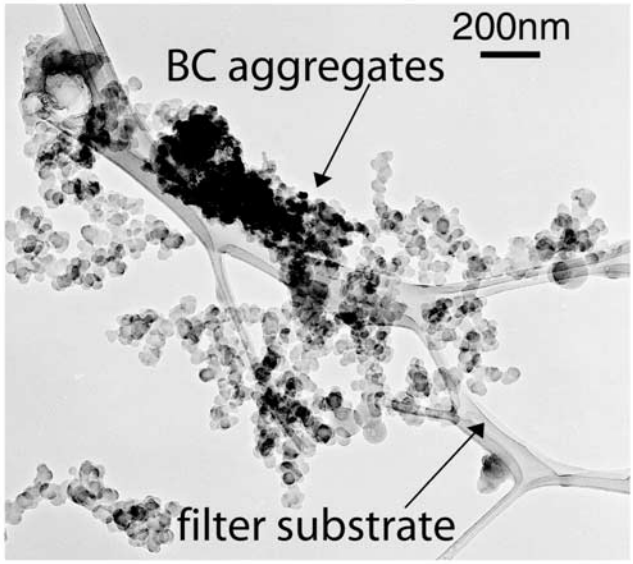

Figure 3. TEM image of BC aggregates in the Timbavati Game Reserve. The age is estimated to be between 12 and 55 minutes subsequent to emission.

difficult to determine owing to the flight pattern performed but the aerosol is estimated to be between 12 and 55 minutes old. As the aerosol ages the chain-like aggregates collapse into densely packed soot clusters [*Martins et al.*, 1998a; *Johnson et al.*, 1991], and the individual components of biomass burning aerosol become internally mixed [*Pósfai et al.*, 2003]. The mass of BC to total aerosol (M_{BC}/M_{tot}) required to match the measurements of the aerosol co-albedo at source and in the aged regional haze was modeled.

[10] The aerosol emitted at source was modeled using an external mixture of the highly absorbing BC with the primarily scattering components. The BC was modeled with the Mie code of *Mishchenko and Travis* [1998], which determines the scattering properties of non-overlapping clusters of spheres. Individual BC spherules were assumed to have a radius of 0.0118 μm and a refractive index of 1.75−0.44 *i* [*WCP*, 1986]. Aggregates consisting of 65 individual BC spheres, with shapes ranging from the chain-like structure associated with fresh aerosol, to the more densely packed clusters associated with the regional haze, were randomly oriented with respect to the incident radiation and their scattering properties modeled. Figure 4 shows the modeled co-albedo at 0.55 μm decreases as the chain-like structure collapses, with a value of 0.97 for the open chain and 0.90 for the packed cluster. This decrease is primarily the result of the particle scattering increasing as the chain collapses. The co-albedo values are much higher than the measurements because only the highly absorbing BC component of the aerosol is modeled. The BC structures in Figure 4 were externally mixed with the scattering component. The scattering component was modeled using Mie calculations with a bi-modal log normal fit to the PCASP size distribution measured in the fresh aerosol and a refractive index of 1.53−0.00 *i* [*Haywood et al.*, 2003a], which results in a $\omega_{0\lambda=0.55\mu m}$ of 1.0. The co-albedo of the composite external mixture of scattering and absorbing aerosol was found to be relatively insensitive to the BC shape (varied by <0.01 with a constant M_{BC}/M_{tot}). Therefore, the changing morphology is unlikely to explain the measured variation in the co-albedo between the fresh and aged aerosol. A M_{BC}/M_{tot} of approximately 12% is required to match the measured co-albedo of 0.16 at source.

[11] The Maxwell-Garnet mixing rule was used to model the M_{BC}/M_{tot} in aged regional haze ($n = 1.54 - 0.018\ i$). The rule adequately represents the internal mixing of BC spheres coated with scattering material, typical of particle structures observed in an aged regional haze in Brazil [*Martins et al.*, 1998a]. The absorbing component was assumed to be BC, with a refractive index of 1.75−0.44 *i* at 0.55 μm [*WCP*, 1986] and a density of 1.7 g cm^{-3} [*Haywood et al.*, 2003a]. The scattering component was assumed to have a refractive index of 1.53−0.00 *i* at 0.55 μm [*Haywood et al.*, 2003a]. By constraining the composite aerosol to have a density of 1.35 g cm^{-3} [*Reid and Hobbs*, 1998], the M_{BC}/M_{tot} is 5.4% for the aged regional haze, which is consistent with measurements using filter mass loadings collected onboard the C-130 aircraft from aged regional haze [*Haywood et al.*, 2003a].

4. Discussion and Conclusions

[12] Airborne measurements over and downwind of an agricultural fire show an increase in $\omega_{0\lambda=0.55\mu m}$ from 0.84 at source to 0.90 in the aged regional haze in 5 hours. *Reid et al.* [1998] find a similar increase of 0.06 in $\omega_{0\lambda=0.55\ \mu m}$ with age from measurements in fresh aerosol and aged regional haze in Brazil. This increase has fundamental implications for climate modeling and could be combined with modeling efforts to better constrain the radiative effect of biomass burning aerosol on regional/global scales. However, the results presented use observations from a single biomass burning fire that may not be representative of the very large number of fires annually in southern Africa.

[13] An attempt was made to obtain closure between the measured $\omega_{0\lambda=0.55\ \mu m}$ and Mie calculations using the varying PCASP size distribution downwind with the fixed refractive index of aged regional haze. The changing size distribution alone cannot account for the increase in $\omega_{0\lambda=0.55\mu m}$. Increasing the imaginary part of the refractive index at the source is required to get consistency. This suggests that it is more important to constrain changes in the effective

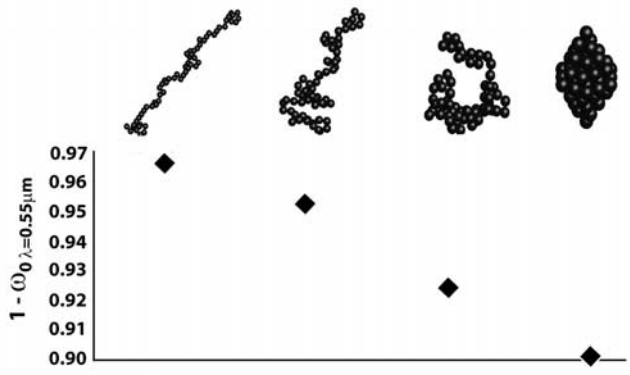

Figure 4. Co-albedo for a variety of BC aggregates, each containing 65 individual spherules.

refractive index than the variation of the size distribution within the plume.

[14] Model results suggest that for $\omega_{0\lambda=0.55\mu m}$ to be consistent with observations the M_{BC}/M_{tot} decreases from approximately 12% at source to 5.4% in the aged haze. The effect of BC morphology on the bulk aerosol absorption is found to be of secondary importance. Therefore, the change in $\omega_{0\lambda=0.55\mu m}$ is likely the result of an increase in the amount of scattering material as the aerosol ages. *Hobbs et al.* [2003] find that subsequent to removing the effect of plume dilution from a separate fire in southern Africa, the number of particles began to increase 10 minutes after emission. This production of new particles can occur by organic carbon (OC) condensing from the gas phase. TEM derived number concentrations of different particle types within a plume during SAFARI 2000 indicate a large increase in the number of tar balls (primarily OC) as the aerosol ages [*Pósfai et al.*, 2003]. *Reid et al.* [1998] observe an increase of 20–45% in aerosol mass concentration from fresh to aged biomass burning aerosol and estimate that between 50–75% of this increase is the result of condensation of volatile organics. However, thermal optical transmission measurements from quartz filters collected onboard the C-130 suggest that the mass of elemental carbon to OC (M_{EC}/M_{OC}) is smaller for fresh aerosol [*Formenti et al.*, 2003], but uncertain due to poor sampling statistics. *Kirchstetter et al.* [2003] also find an increase in M_{BC}/M_{OC} with age but attribute this increase to the biomass burning aerosol mixing with air masses that are characterized by a higher M_{BC}/M_{OC} ratio (e.g., fossil fuel burning). The time scale for this large-scale mixing is likely to be significantly larger than measured/modeled here. Furthermore our model simulations assume that BC is the only significant absorbing component, whereas biomass burning aerosol is likely to contain amounts of absorbing colored organic matter [*Formenti et al.*, 2003]. The apparent discrepancy between our results and those from filter measurements may be elucidated in future measurement campaigns utilising aerosol mass spectrometers.

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